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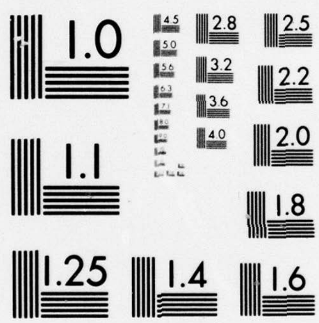
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DEVELOPMENT OF A POWERFUL, WAVELENGTH-TUNABLE INFRARED LASER SYSTEM

FINAL REPORT

for period ending 30 June, 1974

by C. Forbes Dewey, Jr.

U. S. ARMY RESEARCH OFFICE

Contract DAHC04-71-C-0049

FLUID MECHANICS LABORATORY
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The primary objectives of this research program were to develop a high-power, continuously-tunable infrared laser system for the 2 μm - 20 μm region of the spectrum and to advance the fundamental technologies which underlie the practical realization and applications of such a system.

Major achievements of the program include the following:

micrometers 140 300 mt

(1) Nonlinear crystals suitable for phase-matched difference-frequency generation using ruby and dye lasers were investigated. It was demonstrated that continuously tunable phase-matched difference frequencies could be produced from 2 μm t0 22 μm using known nonlinear crystals.

(2) More than thirty dye-solvent combinations suitable for producing laser action in the 710-1100 nm spectral region when pumped with a ruby laser were investigated. We achieved an order-of-magnitude increase in temporal stability of some dyes by using water-surfactant solutions as solvents.

(3) A resonant optoacoustic detection system was conceived and demonstrated in which the absorption of infrared laser sources by trace atmospheric constituents may be measured quantitatively and with high sensitivity.

(4) Room-temperature phase-matched frequency doubling to wavelengths as short as 217.3 nm was achieved in a new nonlinear crystal -- $KB_5O_8 \cdot 4H_2O$. These results extend the use of nonlinear crystals below the previous limit of 235 nm.

(5) We have discovered that periodic phase-matching of nonlinear optical processes can be achieved in crystals exhibiting twin planes. Initial experiments were performed in ZnSe, a cubic crystal not suitable for conventional phase-matching, and we achieved a 100-fold increase in nonlinear optical power by employing a twinned crystal. A single ZnSe crystal produced infrared difference-frequency radiation over the complete spectral range from 3 μm to 21.5 μm.

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I. INTRODUCTION

Our work over the past several years under the present contract has emphasized the production and detection of powerful wavelength-tunable laser sources in the infrared region of the spectrum. The practical application of these techniques is varied, and includes:

- Laser-induced chemical reactions using wavelength-tunable sources
- Isotope separation and other processes requiring high-power laser sources at selected wavelengths
- High-resolution infrared spectroscopy
- Multiple-channel optical communication systems
- Identification and quantification of weak absorption of gaseous molecules, down to 10^{-8} cm^{-1}
- Detection of atmospheric pollutants

The basic physical process which we employ for infrared generation is to produce the difference-frequency between two laser sources in an appropriate nonlinear optical medium. One source is, generally, of fixed frequency (a ruby laser) while the second is continuously tunable (a ruby-pumped dye laser) such that the difference frequency is continuously variable. For the ruby-dye system we employ, it is possible to achieve continuous tuning of the infrared radiation from 2 μm to 22 μm. The broad tunability of this system is not equalled by any other single technique which has been proposed in the literature.

An important aspect of the difference-frequency method is that it is inherently efficient; in principle, 100% of the pump photons of the ruby laser can be converted to infrared photons. Since ruby lasers are among the most powerful laser sources known, the method possesses the potential of producing extremely powerful wavelength-tunable infrared radiation whose power is equal to the ruby laser power times $(\lambda_{ruby}/\lambda_{IR})$, the Stokes shift of the photons produced.

Practical realization of these advantages is impaired by difficulties arising from the nature of the available nonlinear optical materials in which the optical mixing takes place. In addition, severe difficulties have been encountered in the stability of suitable laser dyes. Accordingly, our research has addressed these aspects of the technique. We have made substantial progress in both areas, and are presently embarking on a new approach to the problem of suitable nonlinear materials which could have a substantial impact on this program as well as other nonlinear optical processes.

During the course of our research, we have availed ourselves of the opportunity to exploit two research developments which have arisen. The first is a new method invented under this contract of detecting trace absorption of gases which utilizes a resonant acoustic cell to convert absorbed optical energy to detectable resonant acoustic signals. The second is the discovery of a new family of nonlinear optical crystals capable of generating wavelength-tunable ultraviolet radiation.

II. REVIEW OF ACCOMPLISHMENTS DURING THE CONTRACT

Appendix A summarizes the publications which have resulted from research supported by this contract. Highlights of this work are as follows:

- We have completed our theoretical and experimental investigation of a resonant acoustic chamber for detecting the attenuation of powerful infrared laser beams.

  The system developed under this program is capable of detecting absorptions of approximately 10^{-8} per centimeter, and is applicable to quantitating the weak absorption in the wings of broadened spectral lines as well as detecting the presence of trace atmospheric pollutants at the ppb level.

- The poor temporal stability of some polymethine dyes has inhibited their use in our difference-frequency method of producing wavelength-tunable infrared radiation. We have achieved a factor of 100 increase in the lifetime of these dyes by using water-surfactant solvents.

- We have investigated proustite ($Ag_3 As S_3$) and found that available crystal quality (from RRE) is very poor and it exhibits a very low damage threshold. However, one crystal grown by TYCO withstood approximately 100 MW/cm^2 without damange.

  Our calculations and measurements of infrared spectra suggest that silver thiogallate ($Ag Ga S_2$) should be tunable from 4 - 12 μm, $Li In S_2$ from 3.5 - 9 μm, and $(Zn S)_3 (Ag In S_2)$ from about 7.5 - 16 μm. Progress has been made in characterizing these new crystals.

- We report new results in producing room-temperature phase-matched frequency doubling to 217.3 nm in a new nonlinear crystal - $KB_5O_8 \cdot 4H_2O$. These experiments extend the use of nonlinear crystals below the previous limit of 235 nm achieved in lithium formate monohydrate.

In January of 1974, we embarked on a program to develop suitable periodic structures for use as nonlinear optical media for difference-frequency generation. We performed some initial experiments in which a single ZnSe crystal containing about 150 rotational twins was used to produce difference frequencies continuously tunable from 4 μm to 21 μm.

III. OPTO-ACOUSTIC METHOD OF DETECTION

Research on this phase of our program is reported in Refs. 1, 3, 5, 6, and 8.

We demonstrated acoustic resonant enhancement factors, Q, exceeding 700. The loss mechanisms which determine Q were identified and theoretical calculations confirmed the experimental results.

Subsequent to the publication of our results, several DOD organizations adopted resonant optoacoustic methods in examining the absorption of high-power laser beams in the atmosphere.

Further research to explore the use of wavelength-tunable sources in opto-acoustic spectroscopy would be very useful.

IV. GENERATION OF TUNABLE INFRARED RADIATION IN PERIODIC NONLINEAR CRYSTALS

Conventional nonlinear optical crystals suitable for phase-matched difference-frequency generation have several practical difficulties. Most have relatively modest nonlinear coefficients and high refractive indices, so that the figure of merit, (d^2/n^3), is low. Increases in the optical power density of the mixing beams is often not possible because of the low damage threshold of the crystals. Also, the present scheme utilizing ruby and dye laser radiation requires crystals with large transmission bands from the visible (694.3 nm) to the infrared, further reducing the number of available useful materials. And, finally, the crystals are generally difficult to grow and expensive.

The above facts suggest that progress in the development of suitable conventional phase-matching crystals is possible and worthy of continued support. But alternative approaches should also be vigorously explored.

Recently, we have recognized that certain known infrared-transmitting materials which are not birefringent, and therefore not phase-matchable in the ordinary sense, have sufficiently low dispersion to yield modest coherent lengths for ruby-dye mixing. Of particular interest are ZnS, ZnSe, and ZnTe. The nonlinear figures of merit for these materials, (d^2/n^3), are 75, 160, and 460, respectively. These numbers should be compared to the effective figures of merit for other crystals which we have considered, as listed in Table 1.

The attractiveness of the $\bar{4}3m$ crystals prompted us, in January of 1974, to consider methods in which useful interaction lengths could be obtained. One possibility was the periodic lamellar structures first proposed by Bloembergen (N. Bloembergen, U.S. Patent No. 3,384,433 - 1968); see also J. A. Armstrong et al, Phys. Rev., 127, 1918 (1962)). The formidable fabrication problems attendant to this method were painfully evident.

We succeeded in identifying a naturally-occurring phenomena which exhibits the desirable properties required for producing coherent addition of the optical power

Table 1. Nonlinear Mixing Crystals

Crystal	IR Coverage	Relative Figure of Merit	Damage Threshold (MW/cm^2)
Zn S	2 - 14.5 μm	75	> 100
Zn Se	4 - 22 μm	160	> 100
Zn Te	8 - 52 μm	460	> 100?
$Ag_3 As S_3$ (Proustite)	2 - 12.5 μm	4	5-10 (variable)
$Ag_3 Sb S_3$ (Pyragyrite)	2 - 13.5 μm	4	5-10 (variable)
$Li Nb O_3$	2 - 4.5 μm	5	~50
$Li I O_3$	2 - 6 μm	4	~100
$Li In S_2$	3.5 - 9 μm	5-10?	?
Hg S (Cinnabar)	2 - 15 μm	200	3-5 (variable)
$Ag Ga S_2$ (Silver Thiogallate)	4 - 12 μm	11	10 (variable)
$(Zn S)_3 (Ag In S_2)$	7.5 - 16 μm	16?	High?

radiated by the induced nonlinear polarization. This is rotational twinning. Rotational twins produce a net phase change of polarization by 180° between the two domains. This is more fully described in our first publication on this subject, listed as Reference 10 in the Appendix.

The first experiment demonstrating this effect was performed under this Contract (Reference 10). Tunable infrared radiation was produced from 4 μm to 21.5 μm using our ruby laser-dye laser difference frequency system.

A U. S. Patent, filed under the Contract, has been issued (Reference 12).

V. PHASE-MATCHABLE CRYSTALS FOR INFRARED DIFFERENCE-FREQUENCY GENERATION

Table 1 lists 7 birefringent nonlinear crystals suitable for use in our infrared system. To date, $Li\ NbO_3$, proustite, and $Li\ I\ O_3$ have been successfully employed by ourselves or others who have examined the ruby-dye system, and wavelength tunability has been generated over the 1.5-12.5 μm region.

We examined several of the phase-matchable crystals listed in Table 1. The infrared transmission of $(ZnS)_3(AgInS_2)$ was determined for the first time. Efforts were expended, unsuccessfully, to obtain samples of HgS suitable for phase-matched difference-frequency generation.

VI. ULTRAVIOLET WAVELENGTH-TUNABLE RADIATION

We have successfully produced intense wavelength-tunable ultraviolet radiation in the 217.3 - 234.5 nm region using $KB_5O_8 \cdot 4H_2O$ (potassium pentaborate). This research extends the short wavelength limit of phase-matched frequency doubling in nonlinear crystals below the 235 nm previously reported.

During the final month of the contract period, ammonium pentaborate, a second member of this family, was tested using a nitrogen-pumped dye laser as the fundamental radiation source. These experiments were performed in collaboration with the IBM Watson Research Laboratory. $NH_4B_5O_8 \cdot 4H_2O$ was, on all accounts, less desirable than the potassium homologue.

The new capabilities opened up by high-power, conveniently tunable laser radiation in the 200-230 nm range are numerous. This region includes many atomic and molecular states which are presently unaccessible using previous laser-based sources. This spectral region is of interest with respect to both inorganic and organic molecules. Many biological organisms interact strongly with light of these wavelengths.

VII. IMPROVED LASER DYES AND DYE LASER CAVITIES

Ref. 2, Appendix A, reports on our efforts to increase the temporal stability of laser dyes operating in the near infrared region of the spectrum. Substantial increases in longevity (up to a factor of 100) were observed with the use of aqueous solutions. Prior to our research, water was not used as a solvent for these dyes.

Since the publication of Ref. 2, we have investigated several other polymethine dyes in different solvents. A total of 6 dyes have been investigated, and additional data will be obtained during the course of the difference frequency research of the coming contract period.

Finally, Dr. Hocker has invented (Ref. 10, Appendix A) and successfully demonstrated an integrated dye laser oscillator-amplifier which provides increased coupling between the oscillator cavity and the wavelength-tuning mechanism while providing very high power outputs via an integral amplifier section containing the same dye. The patent application (Ref. 9) was assigned to the U. S. Navy for prosecution.

APPENDIX A

PUBLICATIONS RESULTING FROM RESEARCH

UNDER CONTRACT DAHC04-71-C-0049

1. R. D. Kamm and C. F. Dewey, Jr., "A resonant opto-acoustic technique for pollution monitoring," 1973 IEEE/OSA Conference on Laser Engineering and Applications, May 30 - June 1, 1973.

2. C. E. Hackett and C. F. Dewey, Jr., "Improved temporal stability of polymethine laser dyes in aqueous solutions," IEEE J. Quant. Elect., QE-9, 1119 (November 1973).

3. C. F. Dewey, Jr., R. D. Kamm, and C. E. Hackett, "Acoustic amplifier for detection of atmospheric pollutants," Appl. Phys. Lett., 23, 633 (December 1973).

4. C. F. Dewey, Jr., W. R. Cook, Jr., R. T. Hodgson, and J. J. Wynne, "Frequency doubling in $KB_5O_8 \cdot 4H_2O$ to 217.3 nm," Proc. 8th Int. Quantum Electronics Conf., San Francisco (June 1974).

5. R. D. Kamm, "Detection of weakly absorbing gases using a resonant optoacoustic method," J. Appl. Phys., 47, 3550 (August 1976).

6. C. F. Dewey, Jr., "Optoacoustic spectroscopy," Optical Engineering, 13, 483 (1974).

7. C. F. Dewey, Jr., Laser system for producing wavelength-tunable optical radiation, U.S. Patent 3,731,110 (May 1973).

8. C. F. Dewey, Jr., Resonant opto-acoustic detection system, U.S. Patent 3,938,361 (17 February 1976).

9. L. O. Hocker, Integrated dye laser oscillator-amplifier system, U.S. Patent assigned to U.S. Navy for prosecution (1975).

10. C. F. Dewey, Jr. and L. O. Hocker, "Enhanced nonlinear optical effects in rotationally twinned crystals," Appl. Phys. Lett., 26, 442 (1975).

11. C. F. Dewey, Jr., W. R. Cook, Jr., R. T. Hodgson, and J. J. Wynne, "Frequency doubling in $KB_5O_8 \cdot 4H_2O$ and $NH_4B_5O_8 \cdot 4H_2O$ to 217.3 nm," Appl. Phys. Lett., 26, 714 (1975).

12. C. F. Dewey, Jr., Nonlinear amplifying, U.S. Patent 3,988,593 (October 26, 1976).